

Estimation of historical annual PM_{2.5} exposures for health effects assessment

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Abstract

Epidemiological studies have generally found fine particle metrics such as PM_{2.5} (PM mass less than 2.5 µm in aerodynamic diameter) to be more strongly related to adverse health effects than PM metrics that are not size-fractionated, such as total suspended particulate matter (TSP). The latency of long-term PM exposure effects on health could potentially be investigated using the American Cancer Society Cancer Prevention Study II cohort and other nationwide cohorts. Unfortunately, historical PM_{2.5} data are not available for many past years in most of the US. With the recent introduction of a PM_{2.5} National Ambient Air Quality Standard (NAAQS), fine particulate data is now available through the Environmental Protection Agency's (EPA's) Aerometric Information Retrieval System (AIRS) database from 1999 onwards. Using this nationwide PM_{2.5} data, we have estimated ratios of PM_{2.5}–PM₁₀ (PM mass less than 10 µm in aerodynamic diameter) for more than 100 Metropolitan Statistical Areas (MSAs) in the US. Similarly, using TSP and PM₁₀ data from the late 1980's, when both metrics were measured, we have derived PM₁₀/TSP ratios for hundreds of US MSAs. These MSA-specific PM ratios allow the estimation of historical annual fine particulate concentrations, for as far back as 1972, using available annual TSP or PM₁₀ data. We found mean ratios of PM_{2.5}/PM₁₀ = 0.54 ± 0.14, and PM_{2.5}/TSP = 0.30 ± 0.11. The Inhalable Particle Network (IPN), a database independent of the AIRS database, monitored TSP and PM_{2.5} between 1979–1982. Using a subset of MSAs common to both databases, this dataset has been used to test our hypothesis that MSA-specific mass ratios could be used to estimate PM_{2.5} from PM₁₀ and TSP. Raw IPN TSP–PM_{2.5} concentration correlations for MSAs were non-significant ($R^2 = 0.00$). Using the IPN TSP and our PM_{2.5}/TSP ratios, mean PM_{2.5} estimates for 26 MSAs were found to correlate with the measured IPN PM_{2.5} at $R^2 = 0.43$. These results indicate that it is possible to use MSA-specific PM mass ratios to predict historical annual mean PM_{2.5} exposure levels from past TSP and PM₁₀ measurements. In addition, the MSA-specific ratios were used to estimate nationwide PM_{2.5} concentrations for 1972–2000. These results indicate that considerable progress has been made in reducing U.S. PM_{2.5} levels over the 30 years.

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1. Introduction

Epidemiology studies have found significant associations between exposures to particulate matter (PM) and both mortality and morbidity outcomes. There is growing evidence of health effects as a result of both

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acute and chronic exposures to $PM_{2.5}$. Numerous time-series analyses have focused on $PM_{2.5}$, and found associations between short-term fine PM exposures and the increase in mortality and hospital admissions (e.g. Samet et al., 2000). One recent study from Canada has suggested higher mortality risks to be associated with fine PM, rather than with the coarse PM mode ($PM_{10-2.5}$) (Burnett et al., 2000). This type of study is important for determining how changes in short-term exposures can influence changes in health outcome.

Chronic exposure studies, although fewer in number are very important in assessing the long-term, cumulative impacts of PM. The Harvard Six City and the American Cancer Society (ACS) studies, both prospective cohort studies, have found increased mortality effects associated with long-term exposure to ambient fine PM and sulfates (Dockery et al., 1993; Pope et al., 2002). The ACS study, in particular, found lung cancer, cardiopulmonary and all-cause mortality to be significantly associated with exposure to $PM_{2.5}$, after controlling for individual lifestyle and socioeconomic status indicators. An advantage of this type of cohort study (of large populations followed for multiple years) over the time series analyses, is the availability of covariate information for individual subjects (e.g., occupation, diet, smoking habits). In general, these long-term chronic exposure studies have yielded larger effect size estimates as compared to daily mortality time-series studies, suggesting cumulative PM impacts exist over and above the sum of acute effects (e.g., see US EPA, 2002).

The Harvard Six City and the ACS studies are considered as being two of the cornerstones of PM-health research today, and along with the time series studies have played a pivotal role in the regulation of PM. However, further investigation of the cumulative impacts of $PM_{2.5}$ is necessary to better understand the latency between exposure and health effects, as well as to estimate the potential change in health risk as a result of declining PM exposures. For example, studies on the health benefits of smoking cessation have found mortality rates of former light smokers (who smoked less than 20 cigarettes a day) approach the levels of non-smokers after 10 years (US Department of Health and Human Services, 1990). However, former heavy smokers (who smoked more than 20 cigarettes a day) were still at higher risk after 10 years. This suggests higher exposures are associated with higher mortality risks, as well as benefits of reduced pollution exposures over time. However, the latter may be dependent on the levels and timing of previous exposures. Such exposure–health risk relationships need to be more clearly determined in air pollution studies, and the ACS cohort provides one opportunity to do this.

The ACS Cancer Prevention II Study began in 1982 with an initial population of 1.2 million residing all

across the US. Some half-a-million participants in this study were linked to air pollution monitors to obtain individual exposure estimates on an Metropolitan Statistical Areas (MSA)-specific basis (Pope et al., 1995, 2002). This study used EPA's Inhalable Particle Network (IPN), operational during 1979–1983, along with 1999–2000 Aerometric Information Retrieval System (AIRS) data, to characterize the study population's past and present exposures, respectively. This cohort, with follow-up during 1982–1998, could potentially be used to assess the latency of $PM_{2.5}$ –mortality associations. However, such an analysis would require a complete year-by-year historical fine PM dataset for the period of exposure and follow-up through 1998. The presently limited fine PM exposure data available over this study period presently hinders further temporal investigation of the associated chronic health effects, such as for lag-times of effects.

As a result of the promulgation of a NAAQS $PM_{2.5}$ standard in 1997, fine PM data is now available from 1999 onwards for multiple locations across the US. In fact, many of the published $PM_{2.5}$ time series analyses have been made possible as a result of the availability of this new data. Previous to this time, fine PM data were very limited, with the exception of during the IP Network data system, operational during 1979–1983, which collected TSP and $PM_{2.5}$ data for selected cities (Watson et al., 1981).

To overcome the historical lack of measured $PM_{2.5}$ data, some studies in the 1980s used surrogates of fine PM or devised innovative methods to estimate fine particulate exposures from Hi-Vol TSP and sulfate data, and aerosol extinction coefficients derived from airport visual range observations (e.g. Trijonis, 1983; Ozkaynak et al., 1985). However, these methods are not appropriate for the present ACS analysis, as the use of these methods is not always possible. For example, Trijonis' hybrid methodology applied Hi-Vol TSP, sulfates, lead, and nitrates in a step-wise manner to predict inhalable particles ($IP \leq 15 \mu m$) and Fine Particles ($FP \leq 2.5 \mu m$). In these equations, lead was used as a tracer for vehicular emissions, which no longer holds true, following the removal of lead from gasoline in 1985. The Ozkaynak et al. method of estimation of fine particles using airport visibility data is dependent on individual observers employed by the airports. This data is affected by atmospheric and haze conditions, and by the classification of visibility into broad distance categories, which can limit precision. In addition, airport locations are not usually centrally located and, therefore, pollution measurements made there may be weaker indicators of central population exposures than direct PM measurements.

To respond to these needs, a PM mass ratio method is formulated in this work to estimate historical fine PM exposures from direct PM measurements in each city and year. This method determines urban $PM_{2.5}/TSP$ and $PM_{2.5}/PM_{10}$ ratios for MSAs in the US using available

AIRS TSP (1972 onwards), PM_{10} (1987 onwards) and $PM_{2.5}$ (1998 onwards) data. These computed MSA-specific ratios are subsequently used with the historical annual TSP and PM_{10} data from the same cities in order to estimate annual fine particulate exposures for the specific MSAs where ACS participants have resided. By this approach, nearly 30 years of direct PM measurement data are used in this work to estimate fine particulate exposures of the ACS cohort before and during the study follow-up period (1972–1998). Moreover, in order to independently test the method of fine particle estimation developed in this work, the ratios are applied to the 1979–1983 IP network dataset and tested for fit.

2. Methods

Our approach utilized the variety of PM measurements made in the US MSAs over the last three decades. The EPA promulgated the first National Ambient Air

Quality Standard (NAAQS) for PM in 1971. This regulated ambient Total Suspended Particles (TSP, PM with aerodynamic diameter $\leq \sim 40 \mu m$), and “High Volume” TSP samplers were therefore employed at multiple sites throughout the US. In 1987, these standards were reviewed, and a more health protective standard of thoracic particles (PM_{10}) was introduced. Thereafter, PM_{10} sampling took priority, and the monitoring of TSP gradually declined. It was not until 1997 that the US EPA promulgated a standard for $PM_{2.5}$, resulting in the widespread monitoring of fine particles in over 150 MSAs throughout the US. Therefore, the monitoring of PM has closely followed EPA PM Regulations (see Fig. 1), and TSP, PM_{10} , and $PM_{2.5}$ data are not all available for the entire period from 1972 to present. However, as shown in Fig. 2, there are significant periods where the co-located monitoring of TSP and PM_{10} (1987–1998) and later PM_{10} and $PM_{2.5}$ (1999–2000) occurred for 100s of MSAs, as the state agencies switched from one PM metric to another. These

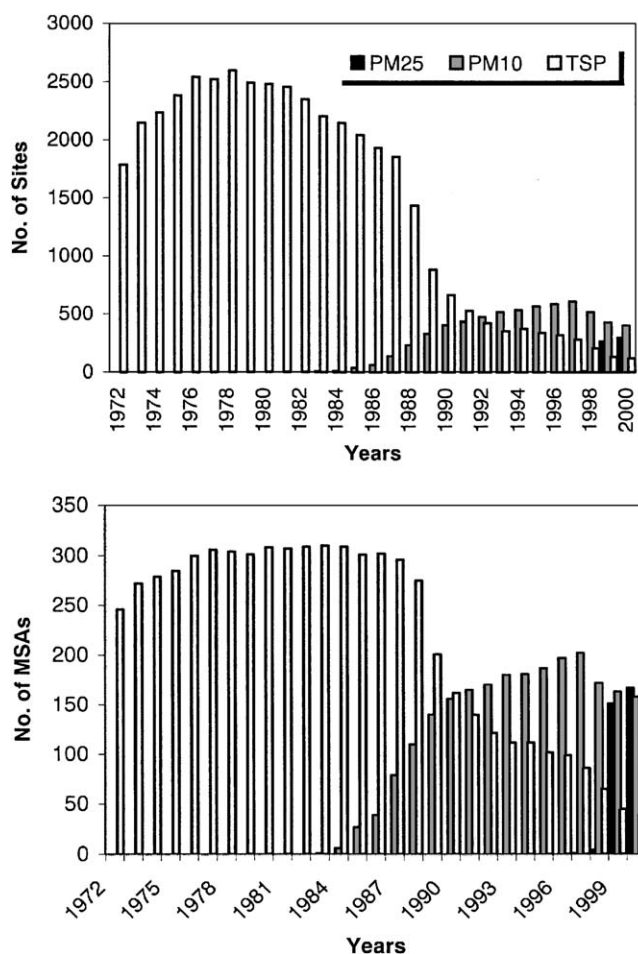


Fig. 1. Number of urban monitoring sites (top) and MSAs with monitoring stations (RIGHT) for PM, in the US during 1972–2000.

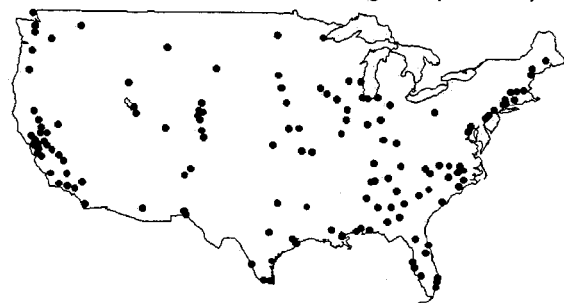
USA: MSA with PM₁₀ and PM_{2.5} Monitoring Sites (1999–2000)USA: MSA with TSP and PM₁₀ Monitoring Sites (1987–1998)

Fig. 2. Periods of overlap in TSP, PM₁₀ and PM_{2.5} monitoring in US MSA's.

overlapping years of PM data, available as quarterly averages from the EPA AIRS database, provide a basis for calculating the MSA-specific fine PM fraction of PM₁₀ and TSP for MSAs across the US. The equations describing these “PM mass ratios” are

$$\text{PM}_{2.5} = b_1 \cdot \text{PM}_{10},$$

$$\text{PM}_{10} = b_2 \cdot \text{TSP}$$

and, therefore, in each MSA

$$\text{PM}_{2.5} = b_1 \cdot b_2 \cdot \text{TSP},$$

where, $b_1 = \text{PM}_{2.5}/\text{PM}_{10}$ ratio, $b_2 = \text{PM}_{10}/\text{TSP}$ ratio, and the product of these ratios ($b_1 \cdot b_2$) provides a fine PM mass fraction of TSP. This method assumes that, while the levels of TSP, PM₁₀, and PM_{2.5} can and do change over time, the relative mix of sources and the resulting PM size distributions stay relatively stable within an MSA. A similar formula was successfully used previously by Trijonis (1983), as a part of a hybrid method to translate Hi-Vol data into estimates of IP and FP (PM_{2.5}).

Quarterly average concentrations of TSP, PM₁₀, and PM_{2.5} for thousands of monitoring sites were retrieved over the Internet from the EPA AIRS database. In order to compute the MSA-specific PM mass ratios, all these data were organized into 2 subsets of PM quarterly data. One consisted of all the monitoring sites where TSP and

PM₁₀ data were both available (i.e. 1987–1998), and the other having all monitoring sites where PM₁₀ and PM_{2.5} data were both available (i.e. 1999–2000). Monitoring sites with at least 50% of observations (i.e. at least 8 observations) per quarter were kept in the dataset. The quarterly data of multiple sites in each MSA were averaged over the multiple years of available PM data, and subsequently averaged across the 4 quarter averages to compute overall paired MSA average values of TSP and PM₁₀, and of PM_{2.5} and PM₁₀ in the two respective subsets. These MSA PM averages were then used in the equations described above in order to derive MSA-specific PM_{2.5}/TSP and PM_{2.5}/PM₁₀ ratios.

TSP and PM_{2.5} data from the IPN from dozens of monitoring sites across the US provide a way to independently test our PM mass ratio method on a set of MSAs common to both datasets. Therefore, the IPN TSP and PM_{2.5} data were organized and averaged using the same procedures described above in order to obtain TSP and PM_{2.5} averages for each MSA. The independently computed PM_{2.5}/TSP ($b_1 \cdot b_2$) mass ratios were then applied to the IPN TSP averages of 26 MSAs to estimate past (1979–1983) PM_{2.5} averages. These PM_{2.5} estimates were then compared with the IPN PM_{2.5} averages. This provided a test of the usefulness of the PM mass ratios in predicting historical PM_{2.5}.

Subsequently, these ratios were applied to the TSP and PM₁₀ annual averages, as computed using the quarterly TSP and PM₁₀ data from the longer AIRS database. During the initial years, when only TSP data was available, the PM_{2.5}/TSP ratio ($b_1 \cdot b_2$) was used to estimate PM_{2.5}. For much of the 1990s, both TSP and PM₁₀ data were available, and therefore an average of the PM_{2.5} estimates derived from each was obtained. For monitors that only measured PM₁₀ (which was the case for some during the latter half of the 1990s) the PM_{2.5}/PM₁₀ ratio was applied. The 1999–2000 observed PM_{2.5} data was averaged with the PM_{2.5} estimations when both were available. For those years when no TSP or PM₁₀ and, therefore, no PM_{2.5} data or estimates were available for an MSA, a generalized additive model of the estimated PM_{2.5}, along with spatial and time smoothing parameters was used, in S-plus 2000, to impute missing PM_{2.5} data (MathSoft Inc., Seattle, WA, USA). A span of 0.04 (about 150 miles) and 7° of freedom (approximately 4 years) were used for this spatial and time smoothing, respectively, for the estimation of missing values.

3. Results

3.1. MSA-specific ratios

MSA-specific PM mass ratios, PM_{2.5}/PM₁₀ and PM_{2.5}/TSP, were computed for 133 MSAs. The

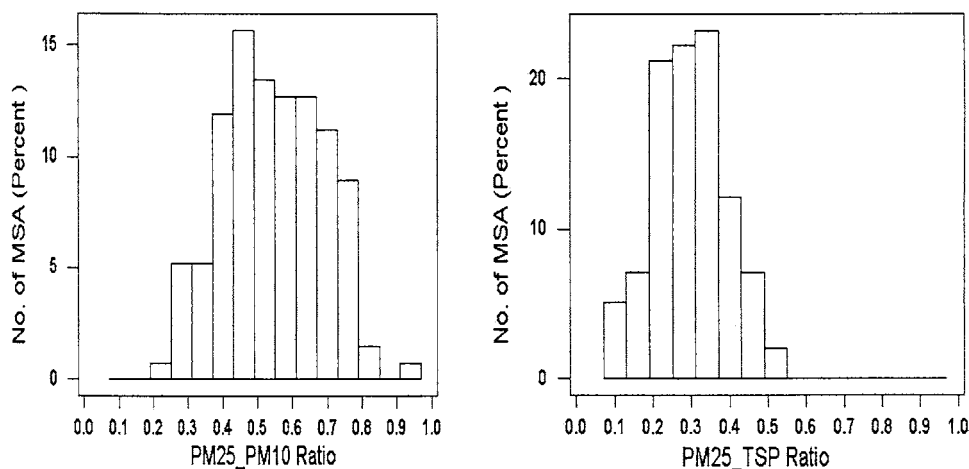


Fig. 3. Frequency distribution of the urban PM_{2.5}/PM₁₀ ratios and urban PM_{2.5}/TSP ratios.

nation-wide ratios appear to be normally distributed (Fig. 3). The mean (\pm standard deviation) of PM_{2.5}/PM₁₀ ratio was found to be 0.54 (\pm 0.14), with a median 0.53, a minimum of 0.20, and a maximum of 0.96. The mean (\pm standard deviation) of the PM_{2.5}/TSP ratio was 0.30 (\pm 0.11), with a mode 0.32, a minimum of 0.10, and a maximum of 0.52. To investigate the variability within an MSA, across sites and between seasons, standard errors were also computed for each MSA-specific ratio (i.e. within MSA variation), yielding PM_{2.5}/TSP standard error of 0.12, and PM_{2.5}/PM₁₀ standard error of 0.14. This indicates that the mass ratio variations over time and space within an MSA can be as large as the “between MSA” variations.

3.2. Dataset of annual PM_{2.5} estimates (1972–2000)

The final dataset consisted of annual PM_{2.5} estimates for 83 MSAs during 1972–2000. In order to estimate the annual fine PM, either annual TSP or PM₁₀ data were required, along with their respective computed MSA-specific PM_{2.5}/TSP or PM_{2.5}/PM₁₀ ratios. A number of MSAs had multiple years of both TSP and PM₁₀ data missing, and therefore, in these cases, the estimation of PM_{2.5} was not possible. Of the original 133 MSAs, 50 MSAs had more than 3 years (between 1972 and 2000) that PM_{2.5} estimates could not be made, and were therefore left out of the final dataset. For the remaining 83, MSAs missing PM_{2.5} values were imputed using a GAM function of the estimated PM_{2.5}, as described in Section 2. Of the 2407 MSA-years of PM_{2.5} estimates, only 2% were filled in this way. The smooth of PM_{2.5} estimations over space and time are shown in Fig. 4 as deviations from the overall mean concentration (16.4 $\mu\text{g}/\text{m}^3$).

3.3. Testing MSA-specific mass ratios on IPN data

PM_{2.5} estimation results for 26 ACS MSAs from the final dataset were matched with IPN MSA observations to allow a comparison of the ratio method PM_{2.5} estimates with actual PM_{2.5} data. The IPN TSP and IPN FP data, for these MSAs, shows no association between these two different PM metrics ($r = 0.04$) (Fig. 5). However, comparison of the IPN FP data for 1979–1983 with the PM_{2.5} estimates from the TSP data averaged for the same period suggests a reasonably strong correlation ($r = 0.65$, $p = 0.0003$). The PM_{2.5} overall average predictions are very slightly overestimated, by approximately 3%. This test indicates that the ratios developed in this work provide a useful approach for estimating past urban fine particulate exposures throughout the US.

In order to make the spatial distributions of the PM mass ratios as well as of the estimated PM_{2.5} levels for the U.S. visually interpretable, the maps in Figs. 6 and 7 were created, respectively. While these maps clearly illustrate the variability between different regions in the U.S., the reader should be aware of the great variability within MSA, which this computation does not incorporate. Therefore, different regions and different periods would have varying levels of confidence associated with the ratios and annual PM_{2.5} estimates presented. For example, as presented above ratio estimates based on TSP have wider confidence intervals compared to those based on PM₁₀, indicating that older PM_{2.5} estimates (based on TSP) are less certain than estimates in more recent years. Furthermore, in Fig. 8, examples of the differing temporal trends within the different MSAs are also shown.

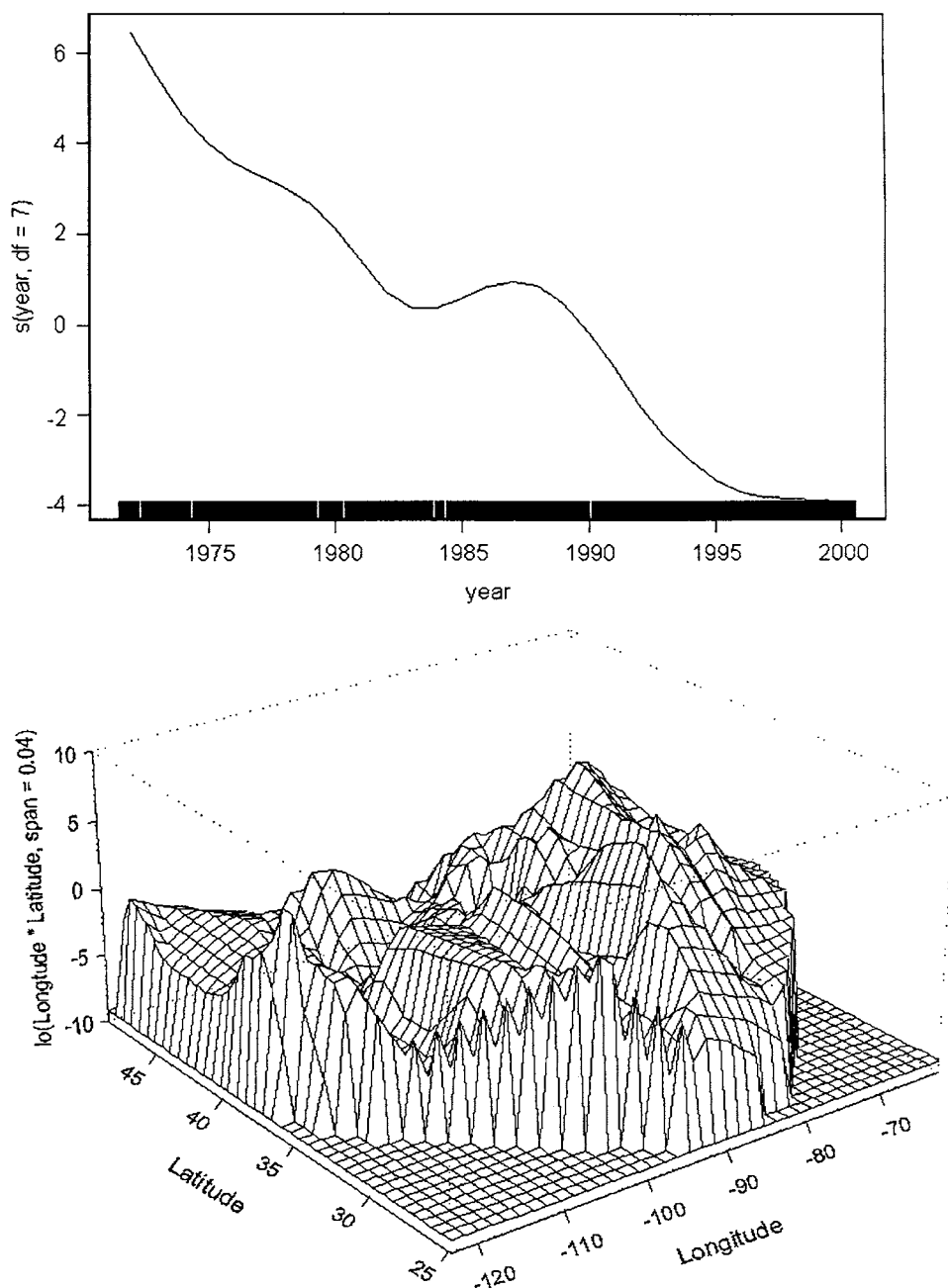


Fig. 4. Time (top) and spatial smooth (bottom) of the estimated US $PM_{2.5}$ data (1972–2000).

4. Discussion and conclusion

In this work, we have estimated two characteristic PM mass ratios for each study MSA: $PM_{2.5}/TSP$ and $PM_{2.5}/PM_{10}$. These MSA-specific ratio estimates were based on multiple sites and multiple years of data, as available

for each MSA. Although the spatial variability of a MSA's PM levels has been incorporated in this approach by averaging the multiple sites available in each MSA, the few years with $PM_{2.5}$ monitoring available to date at these sites is a limitation. Both of the estimated ratios, therefore, rely only on the

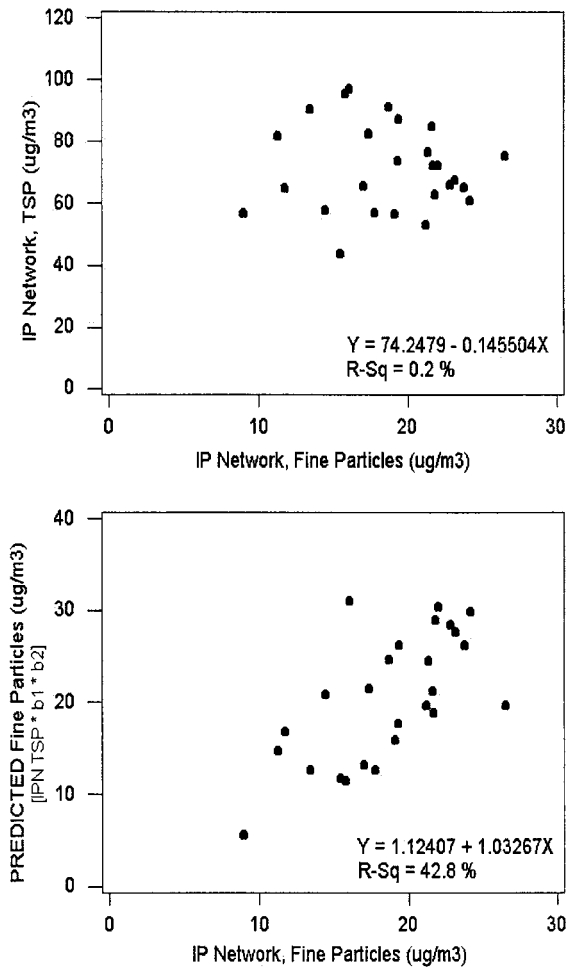


Fig. 5. PM ratios tested on the IPN data; IPN TSP vs. IPNFP (top); IPNFP vs. predicted FP (below).

fine PM fraction data for 1999–2000. This ratio method has the advantage that it allows for the fact that the concentration levels of these pollutants have changed over time. However, the use of such a ratio inherently makes the assumption that, over the last 3 decades, the MSA-specific PM size distributions have remained relatively stable. It is not possible to verify this assumption in all cities due to the lack of historical data between 1985 and 2000. Moreover, information regarding the nationwide changes in PM emission source size distribution is not generally available to validate our assumption. However, the IPN dataset collected during 1979–1983 provided an appropriate means of testing our method on a subset of 26 MSAs that are evenly distributed throughout the US. It also allowed the assumption of the stability of these ratios over time to be checked in this subset of study MSAs. Since this test dataset is for a period of time prior

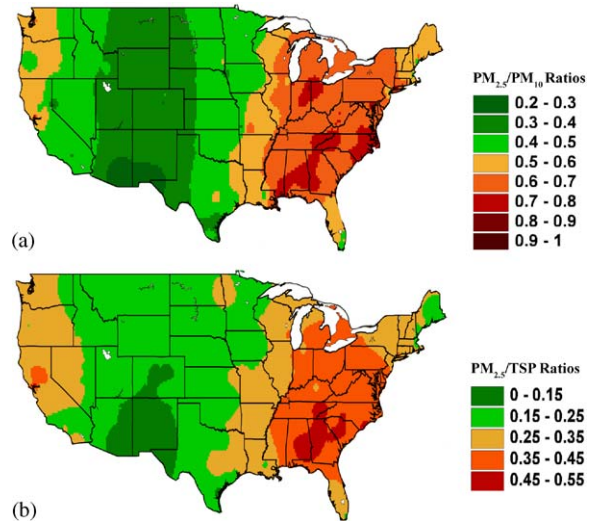


Fig. 6. (a) Distribution of $PM_{2.5}/PM_{10}$ ratios throughout the US. (b) Distribution of $PM_{2.5}/TSP$ ratios throughout the US.

to the years used in the ratio estimation (1987–2000), it represents a severe test of our ratio method. Despite this fact, the ratio-estimated $PM_{2.5}$ and the actual $PM_{2.5}$ concentrations were significantly correlated in these MSAs, and this support the usefulness of this ratio method for the estimation of historical PM exposure in general.

The MSA-specific $PM_{2.5}/PM_{10}$ and $PM_{2.5}/TSP$ ratios show a strong spatial trend across the US, with the north-eastern and eastern parts of the country having among the highest fine mass fractions, and the central US, having much lower fine mass fractions in comparison (see Fig. 6). The higher $PM_{2.5}/PM_{10}$ and $PM_{2.5}/TSP$ ratios in the east are consistent with a greater presence of fine particulate sources (e.g., coal-fired power plants, industry, vehicles and residual oil combustion sources).

The nation-wide declining trends of MSA $PM_{2.5}$ shown in Fig. 7 are reflective of the declining trends of both PM_{10} and TSP. The spatial plot of $PM_{2.5}$ estimates over the three decades, like the mass ratios, indicate higher concentrations being associated with the north-east, the industrial Midwest, and parts of California, while lower concentrations areas are associated with the central US (also see Fig. 4). Interestingly, despite declining levels, the ranking of these MSAs by their $PM_{2.5}$ measured levels has not changed much over time. This was shown previously by the ACS study (Pope et al., 2002) where although $PM_{2.5}$ levels for 1999–2000 AIRS data were on an average some 20% lower than the 1979–1983 IPN data, the levels for these two separate time periods were found to be highly correlated ($r = 0.78$).

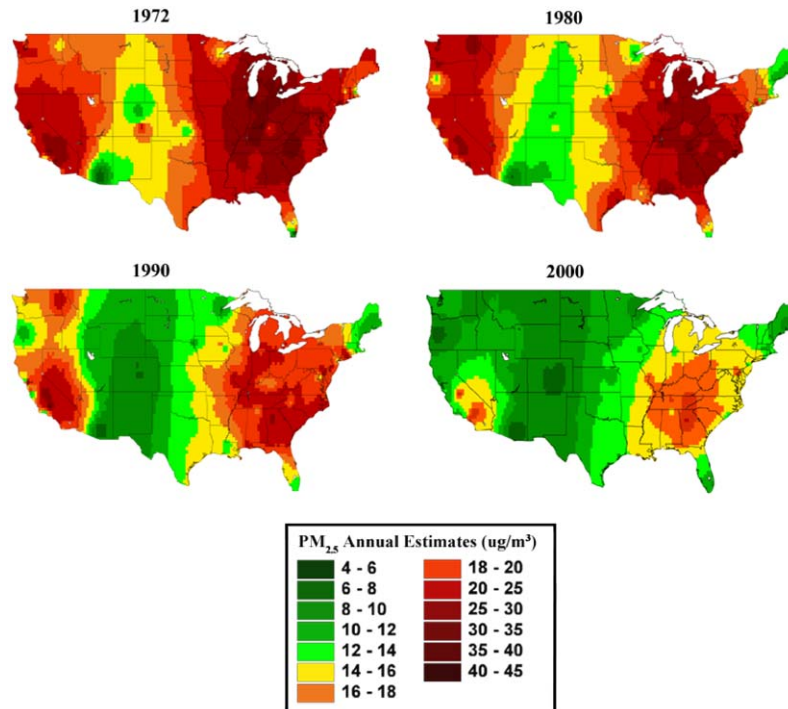


Fig. 7. Declining $PM_{2.5}$ trends over the past three decades for urban areas throughout the US.

It should be noted that there are remaining exposure issues resulting from the use of MSA-level exposures estimates. For example, such exposure estimates do not address the amount of time spent indoors by cohort participants. However, a recent study of personal exposure vs. central site data has suggested that, when estimating health effects from ambient outdoor PM concentrations, the use of central monitoring sites data is appropriate (Mage et al., 1999). Also concentration variations within MSAs are a concern. In future analysis of cumulative, long-term health effects, the “within-MSA” variability of each MSA’s $PM_{2.5}$ levels, as well as the variability in $PM_{2.5}$ levels across the MSAs, may be important to consider. However, given the mobility of people to move within an MSA (e.g., to go to work), the use of an MSA average may represent the most useful estimate of exposures to outdoor air pollution, short of obtaining personal PM monitoring information.

An overview of the $PM_{2.5}$ annual estimates indicate a downward trend for National $PM_{2.5}$ concentrations with a slight peak around 1988 (see Fig. 4). This downward trend is also apparent within individual MSAs, but the declining concentration patterns vary greatly from MSA to MSA (Fig. 8). For reasons similar to those explained above, we are unable to verify the observed time trend for each of these MSAs. To address this problem, error estimates have been provided for these MSA-specific

ratios, in order to better understand the variability within a given MSA. These error estimates could be incorporated into the health effects analysis along with the ratios in future work, potentially providing more accurate confidence bands on health effects estimates. It should be noted that the ratio variations over time and space within the MSAs were found to be roughly as large as the “between MSA” variations. While this would not significantly affect the annual multi-site average comparisons in this work, it could affect health effects assessment of shorter-term exposures (e.g., when seasonal estimates are needed). As more $PM_{2.5}$ data become available, season-specific and sub-MSA region-specific ratios could be computed for use in such studies. Overall, these results indicate that considerable progress has been made in reducing US $PM_{2.5}$ levels over the 30 years.

Based upon the results of this research, the ACS and other long-term studies of air pollution (e.g. the US Veteran Cohort Study (Lipfert et al., 2000)), can use this approach to divide their follow-up time into distinct $PM_{2.5}$ exposure time windows. This would therefore allow the calculation of $PM_{2.5}$ exposure estimates for different time intervals in such cohort epidemiology studies, in turn allowing the investigation of such important issues as time-lag of effects and extent of life-shortening associated with chronic $PM_{2.5}$ exposures.

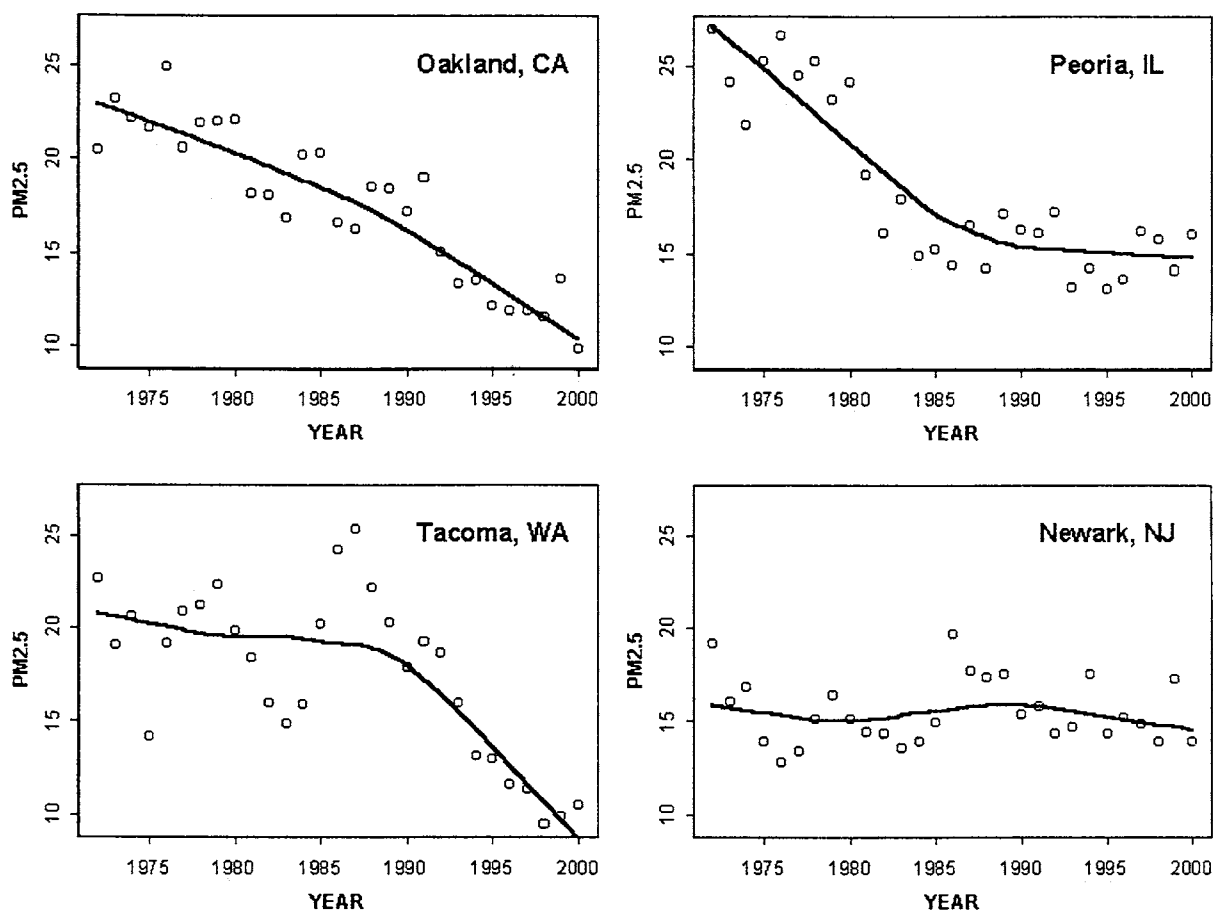


Fig. 8. The pattern of decline in PM_{2.5} varies across MSAs.

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